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⑮ 発明の名称 燃焼用ガスの製造法

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明 細 書

1. 発明の名称

燃焼用ガスの製造法

2. 特許請求の範囲

(1) 好気性菌体が大量に増殖した液を急速に嫌気性状態下におき、水素ガスを発生させることを特徴とする燃焼用ガスの製造法。

(2) 好気性菌体が大量に増殖した液を急速に嫌気性状態下におき、水素ガスを発生させるに際して、光合成細菌を共存させることを特徴とする燃焼用ガスの製造法。

(3) 好気性菌体が大量に増殖した液を急速に嫌気性状態下におき、前記液に光合成細菌を共存させ、水素ガスを発生させた後、さらに嫌気性状態下におき、メタンガスを発生させることを特徴とする燃焼用ガスの製造法。

3. 発明の詳細な説明

〔産業上の利用分野〕

本発明は燃焼用ガスの製造法に関するものである。

更に詳しく述べれば、本発明は好気性菌体が大量に増殖した液から大量の水素ガスとメタンガスを発生させる燃焼用ガスの製造法に関するものである。

そして、本発明は廃水の処理に応用することができるという特徴を有する。また燃焼用ガスの製造過程でできる菌体の沈殿物はそのまま養分として再活用できるという特徴を有する。

〔従来の技術〕

光合成細菌その他の微生物に水素ガスを発生する能力のあることは公知の事実であり、この方面の基礎研究は余りにも多い。しかし、これら基礎研究が多数にのぼつたにも拘らず、その実用化に成功しなかつたのは、水素ガスの発生条件の設定が困難であり、また発生量も少なく、とても経済的生産には結びつかなかつたことに原因があつた。

ガスを採取した後の廃水は BOD 値数 100 ppm 程度にまで浄化されており、この後通常の廃水浄化処理工程、即ち沈殿槽、曝気槽、沈殿槽へ順次移行して放流可能な清水とすることができる。さらに副産物としての菌体沈殿物はそのままで資源として再活用できるものである。ところで、メタン発酵により発生したメタンガスで発電する方法は世界的に実施されてきた技術であるが、この場合にはメタン発酵後の排水が非常に不潔なものであり、そのまま放置すれば、再び汚染を引起すことになるので、どうしても再浄化処理をする必要があつた。しかし折角発生させたエネルギーを再びその浄化処理に消費するというのでは、生産という意味からはマイナスになり、メタン発酵によるエネルギー生産技術はそれ程優れた技術とはいへないものであつたことに鑑みれば、前記した本発明の水素ガス発生後に、メタンガスを発生させる技術は大変優れているといえる。

なお、またメタンガスを発生させた後の菌体沈

殿物がそのまま資源として再活用できるという説明をしたが、実施例 2、4、7、9 及び 11 の水素ガスを発生させた後の菌体沈殿物も、そのまま資源として再活用できるものである。

〔実施例〕

実施例 1.

穀粉工場廃水 (BOD 10,000 ppm) 1000 l を曝気槽に導き、12 時間激しく曝気して、廃水に含まれていた好気性菌体を大量に増殖させた。この好気性菌体が大量に増殖した液を急速に (5 分間で) 嫌気槽へ移行させ、嫌気性状態で 72 時間ゆるやかに攪拌した。その間、発生した燃焼用ガスが嫌気槽上方に溜まるのをガス貯溜タンクに導き貯溜した。発生したガス量は 1 気圧 0.2 m で、その内、水素ガスは 50 %、炭酸ガスは 50 % であつた。

実施例 2

穀粉工場廃水 (BOD 10,000 ppm) 1000 l を曝気槽に導き、別に大量培養した光合成細菌ロドスピリウムカプシラタの培養液を 20 l 添加して、

12 時間激しく曝気して、廃水に含まれていた好気性菌体と別に加えた光合成細菌を大量に増殖させた。この好気性菌体と光合成細菌の増殖した液を急速に (5 分間で) 嫌気槽へ移行させ、嫌気性状態で 72 時間ゆるやかに攪拌した。その間、発生した燃焼用ガスが嫌気槽上方に溜まるのをガス貯溜タンクに導き貯溜した。発生したガス量は 1 気圧 1.2 m で、その内、水素ガスは 80 %、炭酸ガスは 20 % であつた。

実施例 3

アルコール工場廃水 (BOD 10,000 ppm、C/V 比 50) 1000 l を曝気槽に導き 12 時間激しく曝気して、廃水に含まれていた好気性菌体を大量に増殖させた。この好気性菌体が大量に増殖した液を急速に (5 分間で) 嫌気槽へ移行させ、嫌気性状態で 72 時間ゆるやかに攪拌した。その間、発生した燃焼用ガスが嫌気槽上方に溜まるのをガス貯溜タンクに導き貯溜した。発生したガス量は 1 気圧 0.25 m で、その内、水素ガスは 65 %、炭酸ガスは 35 %、

窒素ガスは 5 % であつた。

実施例 4

アルコール工場廃水 (BOD 10,000 ppm、C/V 比 50) 1000 l を曝気槽に導き、別に大量培養した光合成細菌ロドスピリウムカプシラタの培養液を 20 l 添加して、12 時間激しく曝気して、廃水に含まれていた好気性菌体と光合成細菌を大量に増殖させた。この好気性菌体と光合成細菌の増殖した液を急速に (5 分間で) 嫌気槽へ移行させ、嫌気性状態で 72 時間ゆるやかに攪拌した。その間、発生した燃焼用ガスが嫌気槽上方に溜まるのをガス貯溜タンクに導き貯溜した。発生したガス量は 1 気圧 1.5 m で、その内、水素ガスは 70 %、炭酸ガスは 25 %、窒素ガスは 5 % であつた。

実施例 5

豆腐工場廃水 (BOD 10,000 ppm) 1000 l を曝気槽に導き、12 時間激しく曝気して、廃水に含まれていた好気性菌体を大量に増殖させた。この好気性菌体が大量に増殖した液を急速に (5 分間で)

スは50%、窒素ガスは50%であつた。

表 1

実施例 11.

水産加工（魚類解体）工場廃水で、熱処理直後の濃厚廃水（BOD 10,000 ppm、80℃）を熱交換器に通して水温を50℃まで下げる。上記廃水に好気性菌バチルス・メガテリウム、アゾトバクター・ビネランディ、サッカロミセス・セレビシアエを接種し、更に別に大量培養した光合成細菌クロマチエウの培養液を20%添加して、12時間激しく曝気して、廃水に接種した好気性菌体と光合成細菌を大量に増殖させた。この好気性菌体と光合成細菌の増殖した液を急速に（5分間で）嫌気槽へ移行させ、嫌気性状態下で72時間ゆるやかに攪拌した。その間、発生した燃焼用ガスが嫌気槽上方に溜まるのをガス貯留タンクに導き貯留した。発生したガス量は1気圧1.7m³で、その内、水素ガスは65%、炭酸ガスは20%、窒素ガスは15%であつた。

	ガス発生量 (1気圧)	メタンガス (%)	水素ガス (%)	炭酸ガス (%)	窒素ガス (%)
実施例2 の場合	0.6 m ³	60	10	30	0
実施例4 の場合	0.7 m ³	60	9	30	5
実施例7 の場合	0.5 m ³	55	2	35	10
実施例9 の場合	0.55 m ³	50	5	30	15
実施例11 の場合	0.4 m ³	60	5	30	5

〔発明の効果〕

以上説明したように、本発明に係る燃焼用ガスの製造法によれば簡単な方法で、大量の水素ガス、メタンガスが製造でき、エネルギー源の減少著しい今日にあつて見れば、優れたエネルギー生産手段として産業利用価値が高い。

また本発明に係る燃焼用ガスの製造法を実施すれば廃水の浄化が同時に進行しているので、この

廃水の浄化効果に着眼すれば、廃水の浄化施設の運転エネルギーを生産しながら廃水の浄化を実施できるという優れた効果があり、この点からも本発明は産業利用価値が高いといえる。

さらに、また燃焼用ガスの製造過程でできる副産物としての菌体沈殿物はそのまま資源として再活用できる利点があり、この点からも本発明は産業利用価値が高いといえる。

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METHOD FOR PRODUCING COMBUSTIBLE GAS

(I) Claims

1. A method for producing combustible gas, which comprises placing rapidly a liquid in which aerobic microbes have been abundantly proliferated, under anaerobic conditions so that hydrogen is generated.

2. A method for producing combustible gas, which comprises keeping rapidly a liquid in which aerobic microbes have been abundantly proliferated, under anaerobic conditions so that hydrogen is generated in the copresence of photosynthetic bacteria.

3. A method for producing combustible gas, which comprises keeping rapidly a liquid in which aerobic microbes have been abundantly proliferated, under anaerobic conditions, allowing a photosynthetic bacteria to coexist in the liquid, generating hydrogen, and further keeping the said liquid under anaerobic conditions so that methane gas is generated.

(II) On page 2, right upper column, lines 3-18

The aerobic microbe includes *Bacillus megaterium*, *Azotobacter vinelandii*, *Saccharomyces cerevisiae*, *Aspergillus oryzae*, *Candida utilis*, *Rhodotorula rubra*, *Pseudomonas aeruginosa*, *Streptomyces griseus*, and *Escherichia coli*, and there is no particular limitation to the kind thereof. These above-mentioned aerobic microbes are usually contained in various kinds of wastewater.

When a liquid in which aerobic microbes have been abundantly proliferated is rapidly kept under anaerobic conditions, the aerobic microbes are autolysed, thereby causing bubbles coming from the liquid surface. Most of these bubbles are hydrogen gas.

The inventors of the present invention have discovered a new finding that a large amount of hydrogen is generated when a liquid in which aerobic microbes have been proliferated in a large amount is rapidly kept under anaerobic conditions.

(III) Working Examples 1-11

(1) Example 1

Starch factory wastewater (BOD 10,000 ppm) 1000 L was introduced into an aeration tank, and it is vigorously aerated for 12 hours so that aerobic bacterial cells contained in the wastewater were proliferated abundantly. The liquid containing a large amount of the aerobic bacterial cells was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 0.2 m³ of 1 atm. comprising 50% hydrogen gas and 50% carbon dioxide gas.

(2) Example 2

Starch factory wastewater (BOD 10,000 ppm) 1000 L was introduced into an aeration tank. On the other hand, a culture solution 20 L of a photosynthetic bacteria, *Rhodospseudomonas capsulata* which had been abundantly cultured was added to the aeration tank. The mixture was vigorously aerated for 12 hours so that aerobic bacterial cells contained in the wastewater and the photosynthetic bacteria cells were allowed to be abundantly proliferate. The liquid containing these aerobic bacterial cells and photosynthetic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.2

m³ of 1 atm. comprising 80% hydrogen gas and 20% carbon dioxide gas.

(3) Example 3

Alcohol factory wastewater (BOD 10,000 ppm, C/N ratio 50) 1000 L was introduced into an aeration tank, and it is vigorously aerated for 12 hours so that aerobic bacterial cells contained in the wastewater were abundantly proliferated. The liquid containing the aerobic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 0.25 m³ of 1 atm. comprising 65% hydrogen gas, 30% carbon dioxide gas and 5% nitrogen gas.

(4) Example 4

Alcohol factory wastewater (BOD 10,000 ppm, C/N ratio 50) 1000 L was introduced into an aeration tank. On the other hand, a culture solution 20 L of a photosynthetic bacteria, *Rhodospseudomonas capsulata* which had been abundantly cultured was added to the aeration tank. The mixture was vigorously aerated for 12 hours so that aerobic bacterial cells contained in the wastewater and the photosynthetic bacteria cells were allowed to abundantly proliferate. The liquid containing these aerobic bacterial cells and photosynthetic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.5 m³ of 1 atm. comprising 70% hydrogen gas, 25% carbon dioxide gas and 5% nitrogen gas.

(5) Example 5

Tofu factory wastewater (BOD 10,000 ppm) 1000 L was introduced into an aeration tank, and it is vigorously aerated for 12 hours so that aerobic bacterial cells contained in the wastewater were abundantly proliferated. The liquid containing the aerobic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.5 m³ comprising 20% hydrogen gas, 50% carbon dioxide gas and 30% nitrogen gas.

(6) Example 6

Tofu factory wastewater (BOD 5,000 ppm) 1000 L was introduced into an aeration tank. Precipitates were collected from a precipitation tank installed in the processing steps of tofu factory wastewater. These precipitates (5 kg) were added to the above aeration tank, and vigorous aeration was continued for 12 hours so that aerobic bacterial cells contained in the above wastewater and precipitates were allowed to be abundantly proliferate. The liquid containing the aerobic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) to an anaerobic tank and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.5 m³ comprising 20% hydrogen gas, 50% carbon dioxide gas and 30% nitrogen gas.

(7) Example 7

Tofu factory wastewater (BOD 5,000 ppm) 1000 L was introduced into an aeration tank. Precipitates were collected

from a precipitation tank installed in the processing steps of tofu factory wastewater. These precipitates (5 kg) were added to the above aeration tank, and a culture solution 20 L of a photosynthetic bacteria, *Rhodopseudomonas capsulata* which had been abundantly cultured separately was added thereto. Vigorous aeration was continued for 12 hours so that aerobic bacterial cells contained in the above wastewater and the photosynthetic bacteria, *Rhodopseudomonas capsulata* were allowed to abundantly proliferate.

The liquid containing the aerobic bacterial cells and photosynthetic bacterial cells which had been proliferated was quickly (in 5 minutes) to an anaerobic tank and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.7 m³ at 1 atm. comprising 65% hydrogen gas, 20% carbon dioxide gas and 15% nitrogen gas.

(8) Example 8

Chemical fiber factory wastewater (BOD 10,000 ppm, organic acid...90% acetic acid...) 1000 L was introduced into an aeration tank. Activated sludge was collected from the factory, and the activated sludge (5 kg) was added to the above aeration tank. Vigorous aeration was continued for 12 hours so that aerobic bacterial cells contained in the wastewater and activated sludge were allowed to be abundantly proliferated. The liquid containing the aerobic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.2 m³ of 1 atm. comprising 40% hydrogen gas, 45% carbon dioxide gas and 15% nitrogen gas.

(9) Example 9

Chemical fiber factory wastewater (BOD 10,000 ppm, organic acid...90% acetic acid...) 1000 L was introduced into an aeration tank. Activated sludge was collected from the factory, and the activated sludge (5 kg) was added to the above aeration tank. The culture solution 20 L of a photosynthetic bacteria, *Rhodospirillum* which had been abundantly cultured separately was added the aeration tank. Vigorous aeration was continued for 12 hours so that aerobic bacterial cells contained in the wastewater and photosynthetic bacterial cells were allowed to be abundantly proliferated. The liquid containing the aerobic bacterial cells and photosynthetic bacterial cells which had been proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. The generated gas was introduced into a gas storage tank, and then stored therein. The amount of the generated gas was 1.2 m³ of 1 atm. comprising 70% hydrogen gas, 20% carbon dioxide gas and 10% nitrogen gas.

(10) Example 10

Concentrated wastewater (BOD 10,000 ppm, 80°C) obtained after heating wastewater of marine products processing (fish processing) factory was passed through a heat exchanger to lower the water temperature to 30°C. Aerobic bacteria *Bacillus megaterium*, *Azotobacter vinelandii* and *Saccharomyces cerevisiae* were inoculated onto the above wastewater. Vigorous aeration was continued for 12 hours so that the inoculated bacterial cells were allowed to be abundantly proliferated. The liquid containing the aerobic bacterial cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the

anaerobic tank. Then, the generated gas was introduced into a gas storage tank, and stored therein. The amount of the generated gas was 1.5 m³ comprising 20% hydrogen gas, 50% carbon dioxide gas and 30% nitrogen gas.

(11) Example 11

Concentrated wastewater (BOD 10,000 ppm, 80°C) obtained after heating wastewater of marine products processing (fish processing) factory was passed through a heat exchanger to lower the water temperature to 30°C. Aerobic bacteria *Bacillus megaterium*, *Azotobacter vinelandii* and *Saccharomyces cerevisiae* were inoculated onto the above wastewater, and a culture liquid 20 L of photosynthetic bacteria *Chromatium* which had been abundantly proliferated separately was added thereto. Vigorous aeration was continued for 12 hours so that the bacterial cells inoculated in the wastewater and the photosynthetic bacteria were allowed to be abundantly proliferated. The liquid containing the aerobic bacterial cells and the photosynthetic bacteria cells which had been abundantly proliferated was quickly (in 5 minutes) transferred to an anaerobic tank, and stirred slowly under anaerobic conditions for 72 hours, during which time the generated combustible gas gathered in the upper part of the anaerobic tank. Then, the generated gas was introduced into a gas storage tank, and stored therein. The amount of the generated gas was 1.7 m³ of 1 atm. comprising 65% hydrogen gas, 20% carbon dioxide gas and 15% nitrogen gas.

Table 1

	Amount of gas generated (1 atm.)	Methane gas (%)	Hydrogen gas (%)	Carbon dioxide gas (%)	Nitrogen gas (%)
Example 2	0.6 m ³	60	10	30	0
Example 4	0.7 m ³	60	5	30	5
Example 7	0.5 m ³	55	2	33	10
Example 9	0.55 m ³	50	5	30	15
Example 11	0.4 m ³	60	5	30	5

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